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Some calculations and tables on the neutron-induced activity in fallout due to soil and sea-water

by W. F. Greenhalgh

Summary

These tables are only intended to be used as background information to be used in reading refs. 17 and 18. The starting point of the calculations was A. W. Klement's report (Ref.1.).

The clean bomb

Regardless of whether such a weapon is probable or possible, a clean bomb is taken to be one in which 10% of the energy release is due to fission and 90% due to fusion. It is estimated that a clean bomb produces thirteen times the number of neutrons produced by a pure fission bomb of the same yield.

Assumptions

Fission: 1.4 neutrons available for absorption per U238 fission on the average. The total energy release per fission is 190 MeV.

Fusion: Number of neutrons available for absorption 1.5, say, per fusion on the average. Energy per fusion is 15 MeV (say). See ENW April 1962. p.22.

Ratio of numbers of neutrons produced

Considering two explosions of the same yield, one all fission and the other all fusion,

$$\frac{\text{No. of fusion neutrons}}{\text{No. of fission neutrons}} = \frac{190}{15} \times \frac{1.5}{1.4}$$

$$= 14$$

Consider now a 10% fission weapon.

$$\frac{\text{No. of neutrons}}{\text{No. of neutrons if 100\% fission}} = \frac{(0.1 \times 1) + (0.9 \times 14)}{1}$$

$$= 13$$

Effect on activities in fallout

It follows that the ratio $\frac{\text{activity of isotope}}{\text{activity of F.Ps.}}$, worked out on the assumption of a 100% fission weapon, can be increased by a factor of 130.

Taking Na at 26 hours after burst as a worst case and using the NRDL fission product decay law normalised at H + 1, the total soil activity at this time can change from

$7{,}150 + 2{,}000 = 9{,}150$ MeV/sec/MT $\div 3.7 \times 10^{16}$

with a 100% fission bomb to

$715 + 26{,}000 = 26{,}715$ MeV/sec/MT $\div 3.7 \times 10^{16}$

with a 10% fission weapon.

With a 10% fission weapon, the sodium can predominate over the fission products from about H + $\frac{1}{2}$ to about H + 140. Initially the decay would be slower than $t^{-1.2}$ up to H + 26, and after H + 26 would be faster than $t^{-1.2}$.

Calculation of neutron-induced activity in other cases

The neutron-induced activity cannot be calculated without knowing the complete chemical composition of the medium in which the neutrons are absorbed. The tables might be of help in calculating a possible upper limit to the activity. In particular hydrogen, because of its large absorption cross-section, is important in such things as the moisture content of soils, and the water of crystallisation in mineral rocks.

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SAN 59 6/15/2

Location \ Element	Percentage by weight			
	Na	Mn	K	Al
Earth's crust*, world average Refs. 7 and 12	2.83	0.100	2.59	8.13
Earth's crust*, world average Ref. 2	2.8	0.075	2.6	7.8
Liberia Africa Ref. 3	-	0.008	-	7.89
Nevada desert " "	1.30	0.04	2.70	6.90
Lava clay, Hawaii Ref. 3	0.16	2.94	0.88	18.79
Beach sand Pensacola, Florida Ref. 3	0.001	-	-	0.006
Nevada Test Site soil Ref. 1	1.60	0.04	2.50	6.80
British Isles' soil Ref. 12	?	0.02 to 0.30	?	?
English chalk soil Ref. 11	?	?	?	2.7
Welsh shaly subsoil Ref. 11	?	?	?	10.6

*The earth's crust excludes the oceans and the atmosphere.

Table 1. Composition of soils.

ROCK \ ELEMENT	Na	Mn	K	Al
Average igneous rock Refs.10 & 12	2.8	?	2.6	8.1
Average shale Refs.10 & 12	0.96	?	2.7	8.1
Average sandstone Ref. 10	0.33	?	1.1	2.5
Average sandstone (BRS)	0.74	?	0.83	3.2
Average limestone Ref. 10 & BRS	0.04	0.035	0.27	0.43
Average sediment Ref. 10	0.84	?	2.4	7.1
Quartzite Ref. 12	?	?	?	1.5
Feldspar and feldspathoid Ref. 12	?	?	?	13.5
Kaolinite ,,	?	?	?	21.0
Clay sediments Ref. 12*	0.5	0.05	?	?
Upper lithosphere Ref. 12	?	?	2.5	?
Average granite (BRS)	2.7	0.18	3.3	8.5
Average dolerite (BRS)	2.6	0.09	1.2	8.5

*And taking into account data supplied in private communication by Petrographical Dept. of the Geological Survey and Museum, S. Kensington.

Table 2. Composition of rocks and minerals.

	% by weight
Gravel	28
Sand	27
Bricks	24
Cement	13
Lime	2.9
Timber	2.5
Plaster and plasterboard	1.3
Steel	0.8
Iron	0.53
Copper	0.15
Glass	0.13
Lead	0.09
Aluminium	0.004
TOTAL	100.404

Table 3. Average percentage occurrence of building materials in British buildings.

Material \ Element	Percentage by weight			
	Na	Mn	K	Al
Buildings as a whole (1)	0.16	0.033	0.71	3.0
Gravel (2)	1.8	0.10	1.6	5.8
Sand (3)	0.7	?	0.8	3.2
Bricks (4)	0.56	0.077	2.6	10.7
Cement (Portland)	0.15	0.05	0.58	2.9
Lime	0.05	0.035	0.17	0.4
Timber				
Plaster and plasterboard	0.07	?	0.25	0.7
Steel	?	0.60	?	?
Iron (cast)	?	0.40	?	?
Glass	11	0.035	?	0.6

See table 3 for the average percentage occurrence of these materials in British buildings.

(1) Information supplied by BRS. Based on annual consumption of building materials in UK in 1959.

(2) 'Gravel' is the same as 'ballast' used in concrete making. The standard '1 : 2 : 4' concrete has the following approximate percentage composition by weight.

Cement	Sand	Ballast	Water	
11	28	54	7	%

Limestone, granite and dolerite are all used as gravel according to locality. The composition given is simply the unweighted average for these three.

(3) No analyses of sand are available. The values are for sandstone given by BRS.

(4) BRS data. All types of bricks. Clay analyses supplied in a private communication by the Petrographical Dept. of the Geological Survey and Museum, South Kensington gave averages: Na 0.36% and Mn 0.054%.

Table 4. Chemical composition of British buildings and building materials.

Time after burst	γ power MeV/sec/MT ÷ 3.7×10^{16}
1 hour	525,000
7 hours	50,000
1 day	11,700
2 days	5,000
1 week	1,120
1 month	210
1 year	3.5

Fission fraction 100%.

750,000 gamma Mega Curies at H + 1 (NRDL).

$t^{-1.2}$ law merging into NRDL curve at 100 days after burst.

Average quantum energy 0.70 MeV per disintegration, independent of time.

Table 5. Activity of fission products.

Nuclide	γ power MeV/sec/MT ÷ 3.7×10^{16}			
	0	1 hour	1 day	1 month
(F.P's.)	-	525,000	11,700	210
Na^{24}	6,700	5,900	2,250	-
Al^{28}	149,000	28	-	-
K^{42}	61	58	15	-
Mn^{56}	3,700	2,900	6.7	-

It is assumed that 10^{26} neutrons/MT are absorbed in the soil.

Table 6. Activity of nuclides from soil.

Nuclide	% by wt. of element in soil	Av. quantum energy per disintegration	Half-life	Time of max. activity of isotope / activity of F.P's.	Max. ratio activity of isotope / activity of F.P's.
Na^{24}	1.6 %	4.14 MeV	15 hours	26 hours	19%
Al^{28}	6.8 %	1.78 "	2.3 mins.	4 mins.	-
K^{42}	2.5 %	0.35 "	12.4 hours	21.5 hours	0.14 "
Mn^{56}	0.04 %	1.76 "	2.6 hours	4.5 hours	1.3 "

Table 7. Properties and occurrence of nuclides from soil.

Nuclide	γ power MeV/sec/MT ÷ 3.7×10^{16}			
	0	1 hour	1 day	1 month
(F.P's)	-	525,000	11,700	210
Na^{24}	1,990	1,890	658	-
Cl^{38}	4,980	1,650	-	-
$Na^{24} + Cl^{38}$	6,970	3,540	658	-

It is assumed that 10^{26} neutrons/MT are absorbed in the sea water.

Table 8. Activity of nuclides from sea water.

Nuclide	% by wt. of element in sea water Ref. 8	Av. quantum energy per disintegration	Half-life	Time of max: activity of isotope / activity of F.P's.	Max. ratio: activity of isotope / activity of F.P's.
Na^{24}	1.06	4.14 MeV	15 hours	26 hours	5.6%
Cl^{38}	1.90	1.50 "	37 mins.	64 mins.	0.33%

Table 9. Properties and occurrence of nuclides from sea water.

Time after burst	γ POWER (MeV/sec/MT ÷ 3.7 x 10^{16}) F.P's. 100% Fission	Na^{24} 1.6% in soil	Mn^{56} 0.04% in soil	Na + Mn	13 x (Na + Mn)	0.1 x F.P's.	13 x (Na + Mn) + 0.1 x (F.P's.)
0.5 hrs	1,200,000	6,500	3,200	9,700	126,000	120,000	246,000
1	513,000	6,400	2,800	9,200	119,500	51,300	171,000
2	230,000	6,000	2,150	8,150	106,000	23,000	129,000
3	145,000	5,800	1,650	7,450	96,700	14,500	111,000
4	98,000	5,500	1,250	6,750	87,700	9,800	97,000
5	78,000	5,250	970	6,220	80,800	7,800	89,000
6	61,000	5,000	740	5,740	74,600	6,100	81,000
7	50,000	4,800	570	5,370	69,800	5,000	75,000
8	42,500	4,600	430	5,030	65,300	4,250	70,000
9	37,000	4,400	330	4,730	61,400	3,700	65,000
10	33,000	4,200	260	4,460	58,000	3,300	61,300
12	26,500	3,800	150	3,950	51,300	2,650	53,950
15	20,000	3,300	67	3,367	43,800	2,000	45,800

The soil contains Na 1.6% and Mn 0.04% by weight.
Neutrons absorbed in soil 1.3 x 10^{27} /MT.

Table 10. Fallout from a clean bomb on soil. (See Fig. 5.)

Time after burst	γ POWER (MeV/sec/MT ÷ 3.7 x 10^{16}) F.P's. 10% Fission	Na^{24} + Cl^{38}	13 x (Na + Cl)	0.1 x (F.P's.) + 13 x (Na + Cl)
0.5	120,000	4,500	58,500	178,000
1	52,000	3,500	45,500	97,000
2	23,000	2,300	29,900	53,000
3	14,500	1,850	24,100	38,600
4	9,800	1,700	22,100	31,900
5	7,800	1,570	20,400	28,200
6	6,100	1,500	19,500	25,600
7	5,000	1,430	18,600	23,600
8	4,250	1,360	17,700	21,900
9	3,700	1,300	16,900	20,600
10	3,300	1,250	16,200	19,500
12	2,650	1,130	14,700	17,350
15	2,000	980	12,750	14,750

Seawater contains Na 1.06% and Cl 1.90% by weight (Ref. 8)
Neutrons absorbed in seawater 1.3 x 10^{27} /MT

Table 11. Fallout from a clean seaburst. (See Fig. 5.)

	Fraction by mass
Na in Na_2O	0.74
Mn in Mn_2O_3	0.70
K in K_2O	0.83
Al in Al_2O_3	0.53

Table 12. Percentage by mass of elements in compounds.

	Suburban terraced two-storey house estate - fairly open Ealing, London	City centre Liverpool 6-storey buildings
Fraction of area occupied by buildings	0.20	0.55
Mass per unit area of building $\left\{\text{tons/ft}^2\right\}$	0.16 to 0.24	0.45 to 0.60
Mass per unit area of territory $\left\{\text{tons/ft}^2\right\}$	0.032 to 0.048	0.25 to 0.33

Table 13. Mass of buildings per unit plan area in cities.

Fig. 1. COMPARISON OF FISSION PRODUCT DECAY LAWS.

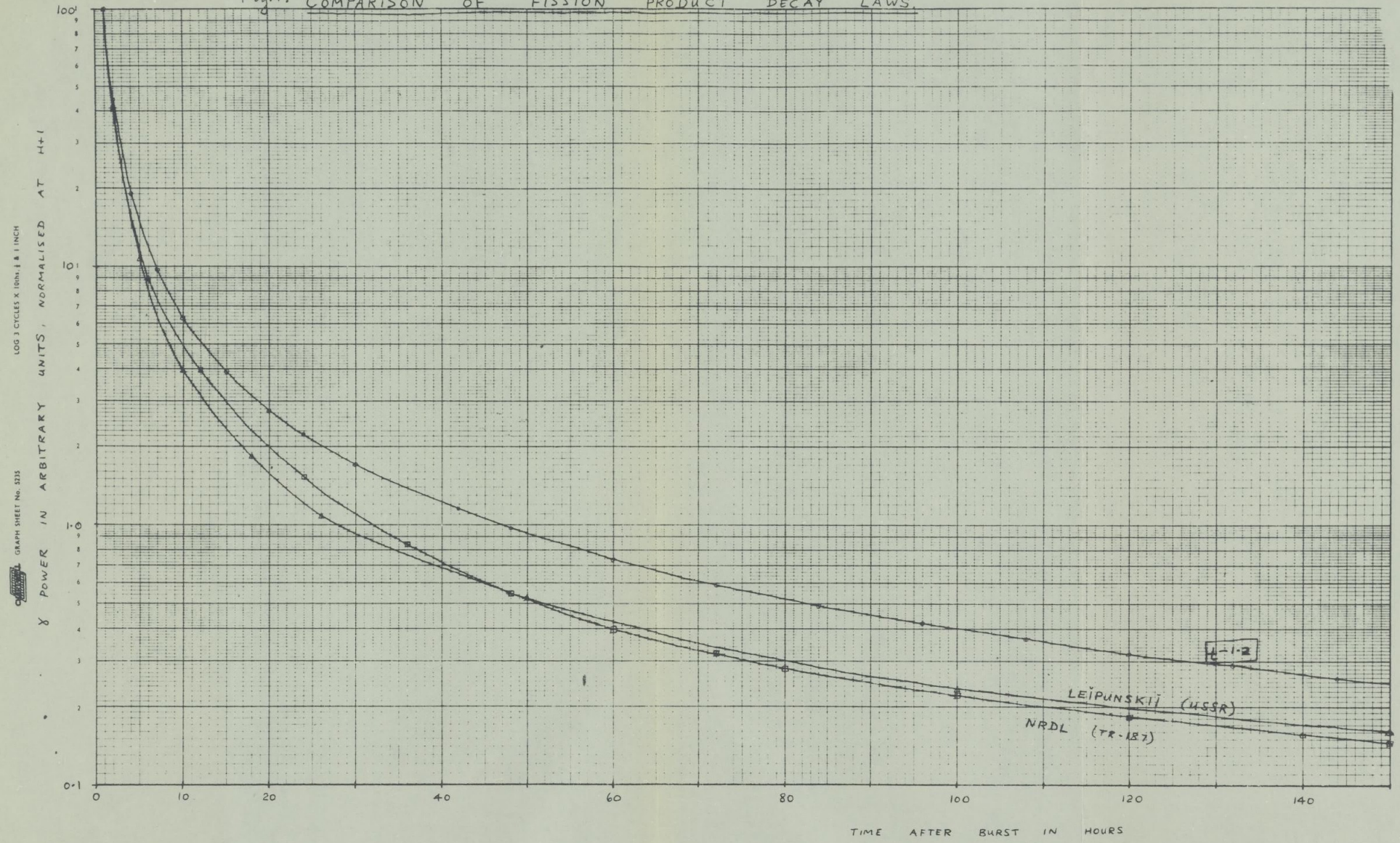
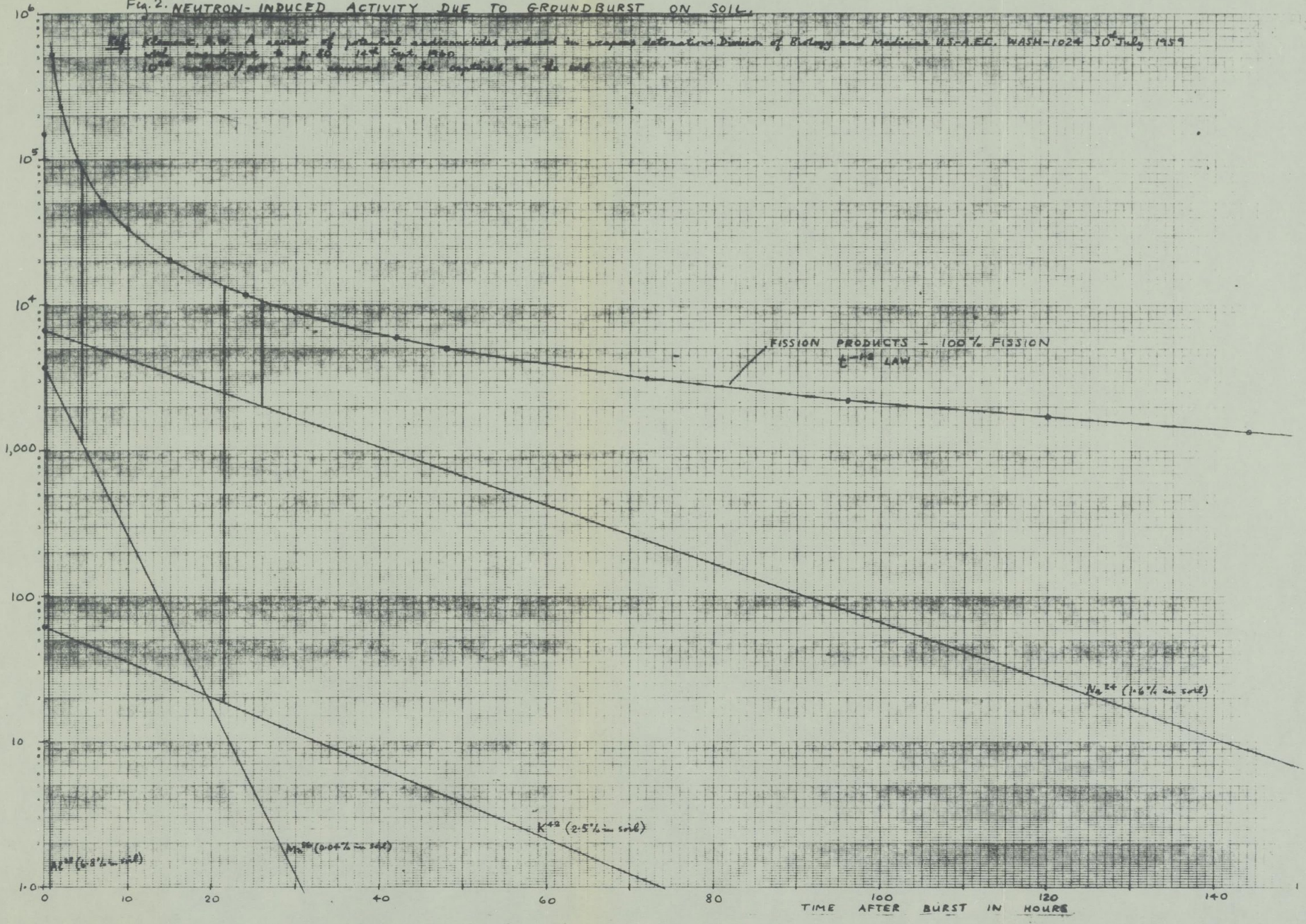


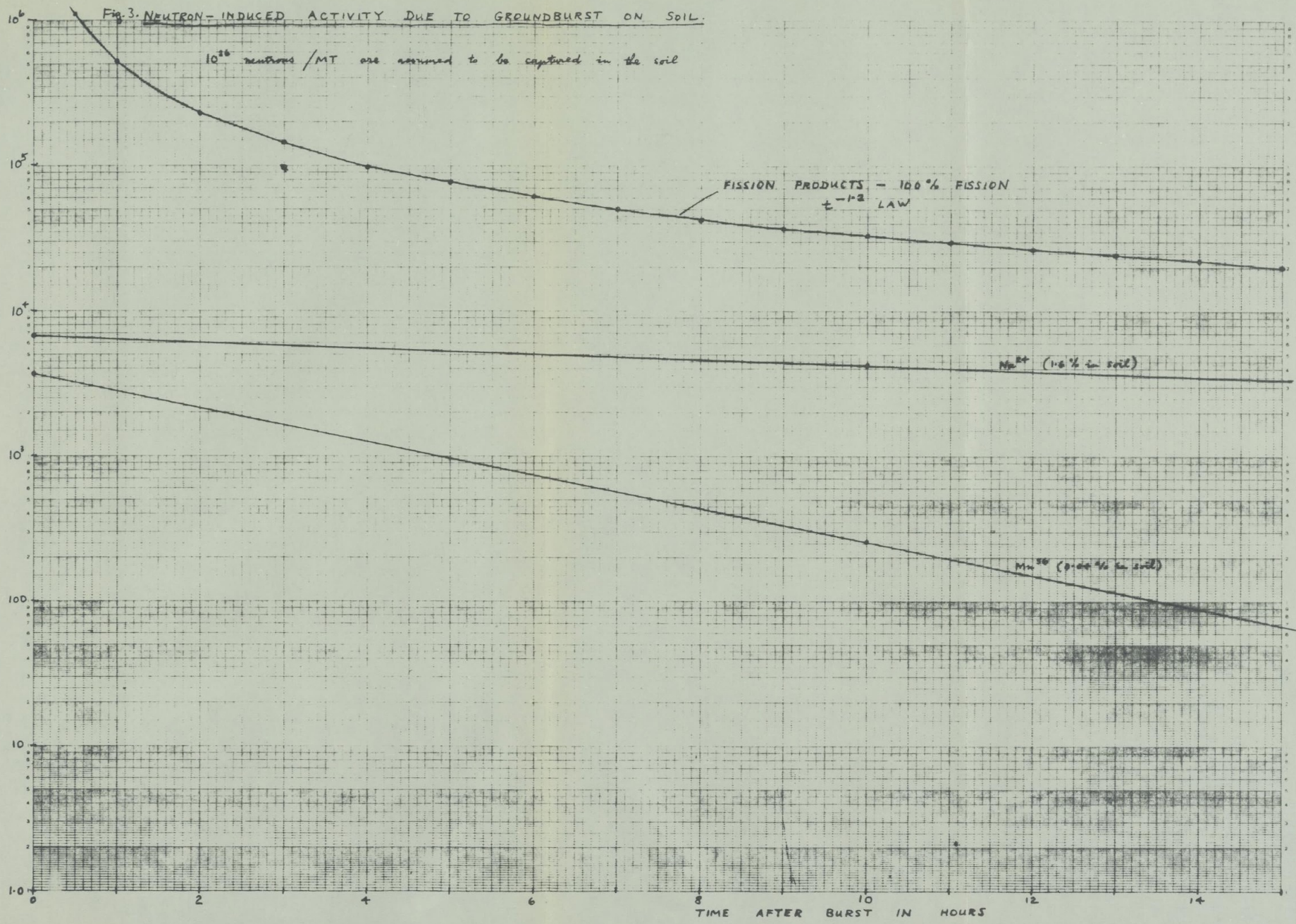
Fig. 2. NEUTRON-INDUCED ACTIVITY DUE TO GROUNDBURST ON SOIL.

Ref: Klement, A.W. A review of published radionuclides produced in weapons detonations. Division of Biology and Medicine U.S.A.E.C. WASH-1024 30th July 1959
[illegible] 14th Sept. 1960.
10^{26} [illegible] per ton equivalent to 22 megatons in the soil



DATA SHEET No. 47. LOGARITHMIC × 10 INS (6 CYCLES)

Fig. 3. NEUTRON-INDUCED ACTIVITY DUE TO GROUNDBURST ON SOIL.



DATA SHEET NO 474 LOGARITHMIC X TIMES (6 CYCLES)

Fig.4. NEUTRON-INDUCED ACTIVITY DUE TO [redacted] SEA BURST.

Ref. Klement, A.W. A review of potential radionuclides produced in weapons detonations Division of Biology and Medicine U.S.-A.E.C. WASH-1024 30th July 1959 with amendment to p. 80 14th Sept. 1960
10^{22} neutrons/MT were assumed to be captured in the sea-water

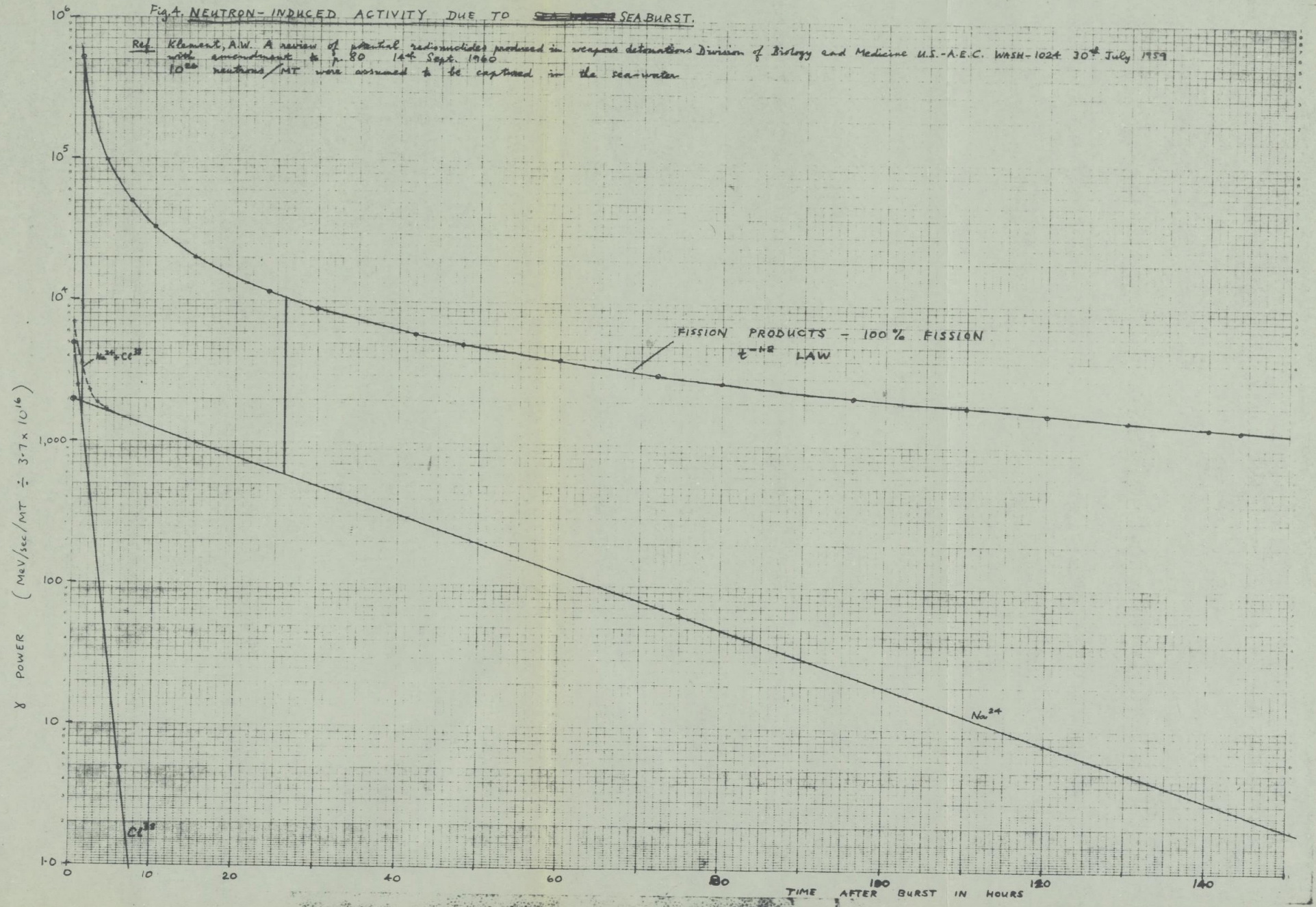


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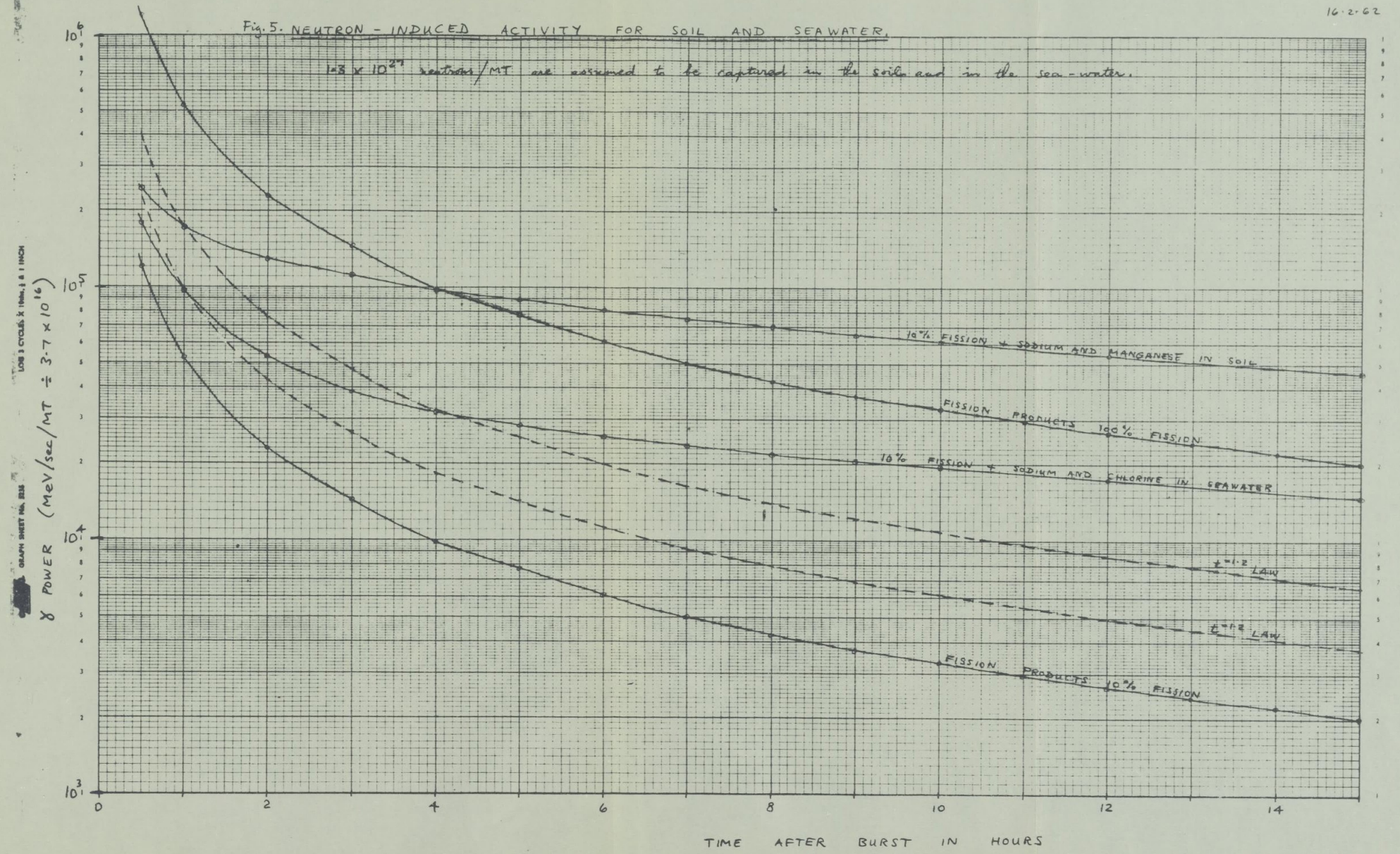
Fig. 5. NEUTRON - INDUCED ACTIVITY FOR SOIL AND SEAWATER.

1.8×10^{27} neutrons/MT are assumed to be captured in the soil and in the sea-water.



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"Fall-out and Radiological Countermeasures Vol I"

Prepared by Carl F. Miller

for OCD, Washington and issued January 1963

Comments and explanations for CD planning

by J. McAulay

Introduction

This volume covers such an extensive field and is so condensed that it is virtually an encyclopaedia on fall-out. Because of this and the many intricate relationships between the parameters used and also because of the "round about" arguments, the report is not easy to read and it is difficult to extract information of direct value for CD planning. The difficulty is increased by the inadequacy of the list of definitions of symbols and particularly by the many errors in cross references apparently not corrected when the material was re-arranged.

Unfortunately also, Miller draws his data solely from unclassified reports and much of it from the 1957 Edition of the "Effects of Nuclear Weapons" (ENW), although a later edition was published in May 1962. Consequently lengthy and involved arguments are often used to establish relationships between some of the parameters and it often requires a careful study of widely separated parts of the report before one can distinguish between tabulated or graphical data based on theory and on experiment or on some mixture of both.

The purpose of the present note might be taken as an attempt to interpret Carl Miller but at least it is hoped to indicate those techniques and results which are likely to be of greatest value for C.D. planning.

Miller's Main Objective

This is to devise a mathematical model of the fireball-cloud, which will take account of the thermodynamic condensation processes in the expanding and cooling fireball and which will also describe adequately the physical, chemical and radiological characteristics of fall-out particles at any point -

(a) within that part of the contaminated area in which the internal gamma hazard is of primary concern in civil defence

(b) in the more extensive area in which the predominant hazard could be that due to the ingestion through the food chain of fission products preferentially retained in the body (e.g. radioactive Sr and I); the biological availability to plants and animals in this area is therefore also a major part of the objective.

Miller has achieved the first part of his objective to a limited extent i.e. the reproduction of idealised dose-rate contours appropriate to the circumstances of a nuclear detonation at weapon trials but he has found that the available experimental data from weapon trials are quite inadequate for the second part of his objective.

Miller's simplified model based on Anderson's dynamic cloud model

The outstanding value of this report lies in the method used by Miller to adapt a mathematical model for reproducing the geometry of the fireball (pp. 141, 142 & 147) and of the rising cloud and stem (pp. 207-209) and pp. 211-214). This model was originally proposed by A.D. Anderson

in USNRDL Report 249 dated 1958. In this model the hemispherical fireball of a groundburst becomes a sphere as it leaves ground level; it then expands adiabatically as it rises to culminate in a stable oblate spheroidal cloud after the toroidal motion has ceased. The stem is the volume swept out by the rising, expanding fireball and fall-out of the larger particles starts as the fireball rises. Of the particles carried to the top by toroidal motion, the heavier ones are thrown at high speed downwards from the periphery of the cloud (p.217), contributing to the high intensity peak from the stem near ground zero and usually separated by a skip distance from the ridge of high intensity caused by particles that fall from the stabilised cloud.

Miller uses the Anderson cloud model, in modified form, to describe the descent of fall-out particles and to calculate for any selected location, The particle size, the mass deposit per unit area and the associated activity of the particles, from specific nuclear detonations of known depth or height of burst, of known fission and total yield and of known type of fissile material (see fission type relationships in Fig. 3.5 p.185).

The name "simplified model" may be appropriate in principle but it is questionable in practice. Nevertheless, as Miller claims, his simplified model does appear to offer a better reproduction of fall-out patterns from nuclear weapon trials than any of the other models devised to-date (see comparison of model predictions pp. 293-298).

The modification of the Anderson model consists in the separate treatment of the fall-out from the stem and from the cloud and in the use of a schematic intensity profile along the central "hot" lines of an idealised fall-out pattern. Fig. 5.1 p.233 illustrates the double humps of high intensity and the selected nine radiation intensity reference points along the "hot" line from the upwind edge to the downwind limit. Point 8 corresponds to the maximum pattern half width. Miller also introduces a particle size-location parameter α (see p.207-208 defined as

$$\frac{\text{wind vector}}{\text{particle fall velocity}} \simeq \frac{\text{downwind distance}}{\text{height from which particles fell}}$$

From fall-out patterns at weapon trials, a number of relationships are derived, based on the assumptions listed on pp. 211-212 connecting the various parameters from one reference point to another along the central hot line. Thus relationships are established -

(i) between the parameters α and fission yield W (p.249-250)

(ii) between upwind and downwind distances X and fission yield, sometimes through other parameters which can be related exclusively to fission yield, such as Rs the radius of the fireball as it leaves the ground and stem variables for height $\bar{z}$ and radius a with suffixes relating to position (p.250).

(iii) between radiation intensity* and W, X or α (p.251-252)

(iv) between yield and half width $\frac{1}{8}$ of the stem pattern bulge

(v) between yield and maximum half width $\frac{1}{8}$ of the cloud pattern ellipse (pp. 245-251).

These empirical relationships are summarised in Table 5.3 p.254 for 100% fission yields of 1 KT to 100 MT and a single direction wind speed of

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*The term radiation intensity can have many meanings. Miller uses the term air ionisation -rate at 3 ft. above a contaminated surface as an absolute value in the absence of a man with a measuring instrument: the latter imposes a characteristic shielding factor and Miller shows plots this factor against time during the use of a typical US portable radiac meter (see Fig. 3.9 p.191).

15 mph. The construction of an idealised fall-out pattern for a 1 MT fission yield in a 15 mph wind is illustrated on pp. 255-258 and Fig. 5.2 p.257 shows radiation intensity vs distance plots with distance from point to point.

The effect of different wind speeds is dealt with in pp. 244-245 and consideration is given to the possible crosswind shear effect on pp. 288-293 and Table 5.11 showing the ratio of lateral expansion to downwind travel of the bulge in the cloud pattern for different values of cross windshear Sy in knots/1000 ft. and for fission yields from 1 KT to 100 MT.

The build up of fall-out (TOA to TOC) at 1.87×10^5 ft. (ca. 35 miles) downwind on the central hot line from a 1 MT fission groundburst in a wind speed of 15 mph is tabulated as a function of the particle size - location parameter α on p.261, Table 5.5.

Relation between radiation intensity and mass deposit per unit area in Miller's model

There still remains the problem of relating radiation intensity at any point in a fall-out pattern to the mass deposit of fall-out per unit area at that point. For this purpose Miller introduces the term Mass Contour Ratio at any time t, with the symbol $M_r(t)$, defined on p.301 as the ratio of the mass of fall-out per unit area to the dose-rate at the 3 ft. level measured at H + t hours. The values of $M_r(t)$ and hence of $M_r(1)$ at H + 1 hour are obtained from measurements at weapon trials in units such as mg/sq. ft. per rph at 1 hour.

It has been found that the inverse of the activity in mg/KT (or mg/fission) for equal values of α tends to decrease slightly with yield by a factor $W^{-0.083}$ and Miller accounts for this (p.304 & 321) by introducing a function $f(\alpha)$ in mg/KT or mg/fission where $f(\alpha)$ is proportional to $\left(\text{the dose rate at 1 hour} . M_r(1) . W^{0.083}\right)$. This in turn leads to the general expression (6.16 p.325).

$$M_r(1) = \frac{f(\alpha) . W^{-0.083}}{D . q_x . B \left[r_\alpha(1) . i_{fp}(1) + i_i(1) \right]}$$

where W is the total yield in KT, B is ratio of fission to total yield, q_x is a ground roughness factor and D is the instrument response factor, so that the quantities within the square brackets are absolute air ionisation rates at H + 1 hours, 3 ft. above an ideal uniformly contaminated with actual (fractionated) fission products corresponding to a surface contamination density of one fission per sq. foot. The term $i_{fp}(1)$ is the true contribution which would have been made if all the fission products had reached the ground without fractionation and $i_i(1)$ is the contribution from the induced activity. The term $r_\alpha(1)$ is the fraction of the total fission products per fission actually reaching the ground at locations having the particle size location parameter α. Putting in values of 0.75 for D, 0.75 for q_x, 6.9×10^{-13} and 0.13×10^{-13} (rph at 1 hour/fission/sq. ft.) respectively for $i_{fp}(1)$ and for $i_i(1)$ from p.231, gives equation 6.17 on p.325.

$$M_r(1) = \frac{1.83 \cdot 10^{11} \cdot f(\alpha) \cdot W^{-0.083}}{B\left[r_\alpha(1) + 0.019\right]} \quad \frac{\text{mg/sq. ft.}}{\text{rph at 1 hr.}}$$

Values of α are determined from the mathematical model and from weapon trials data, Miller has plotted $r_\alpha(1)$ vs α in F6.2 p.320 and $f(\alpha)$ vs α in Fig. 6.3 p.324. The latter curve can be treated as three straight sections from which three empirical equations are derived on p.325 relating $f(\alpha)$ to α for values of α from 0.1 to 0.9; from 0.9 to 20 and for $\alpha > 20$: this last equation is guesswork as no experimental data are available for $\alpha > 20$.

Hence for any selected point for which α can be determined the dose-rate at 1 hour and the mass of fall-out deposited per unit area can be calculated.

Since a fall-out producing detonation may occur below or above ground level, Miller introduces (p.327) a mass correction factor K_λ related to λ, the cube root of the yield and the height or depth of burst. Fig. 6.4 p.328 shows K_λ plotted against λ from weapon trials and that it is in good agreement with values calculated from the crater volume.

Miller also deals in detail with fall-out from sea water for which fractionation is less (i.e. fractionation factors are larger, (see Fig. 6.2 p.320 and pp 327-337).

Thermadynamic processes in the Fireball : Fractionation of Fission Products and Induced Activity

A. General Comments

These aspects of nuclear detonations are discussed by Miller in Chapter 3 which is the most speculative and least successful part of the report relating to the development of a mathematical fireball and cloud model. Some general comments may not be out of place, therefore, before the problems of incorporating fractionation and induced activity into the mathematical model are considered.

The lack of adequate experimental data in these fields force Miller to exemplify his main arguments by a number of arbitrary assumptions (pp. 133, 152, 154 and 157). The two most important are (i) that the soil lifted into the fireball has an "ideal" composition $Na_2O \cdot Al_2O_3 \cdot 6\,SiO_2$ with a melting point of 1400°C and other corresponding properties (p.135) and (ii) that "half ~~an~~ energy in the fireball, at the second maximum, is used to heat dissociate and expand the gas molecules from the air and half is used to vaporize, dissociate and expand the gaseous products from the soil" (p.133). This "ideal" soil corresponds to a desert sand so that comparison is possible between calculations, based on Miller's model and those based on weapon trials in Nevada, for the fraction of the energy required to vaporise and melt the soil in the fireball. Miller estimates (p.151) that over the range 1 KT to 100 MT for groundbursts this amounts to between 7.5 and 9.2% compared with an estimate made by C. E. Adams of USNRDL, of about 3% for tower shots and, since a larger fraction of the energy would be used in groundbursts than in tower bursts, the agreement is quite good. Of the crater mass lifted ~~into the fireball~~, Miller estimates (p.154) that the fraction of this soil melted varies from 3.8% to 12% for detonations of 1 KT to 100 MT.

The comparison is more difficult for detonations on coral (p.156-157) where the carrier soil particles consist of CaO with a high melting point of 2580°C and the particles can react with moisture and carbon dioxide in the atmosphere before being deposited on the ground. Nevertheless Miller claims (p.157) that his model gives values for the amount of liquified carrier soil per unit volume of fireball at the soil melting point temperature, $[h(\ell)/V]$ agree with observed data within a factor of 2.

Miller admits on p.146 that the absolute values of the data in many of his Tables and used to calculate the fractionation factors of individual isotopes in fission products are of questionable use for C.D. planning purposes. Nevertheless the general trends and relative values, deduced from his model, for the fission products that could be deposited in fall-out from a nuclear groundburst give a highly informative picture of the processes that cause fractionation and give also valuable approximations of use in C.D. planning.

B. Two stage condensation process in the fireball.

Chapter 3 considers all the thermodynamic processes which occur in the fireball such as, (a) the vaporization, dissociation, recombination, condensation and solidification of the soil material lifted from the crater (eqn. 3.166 p.154), (b) the successive condensation of fission product isotopes or their oxides etc. as the fireball colls to their respective melting points and (c) the large fraction of the fireball energy used in heating and dissociating the oxygen and nitrogen (mainly the former, pp. 122-123) occluded in the rising and expanding fireball.

The fission product condensation process is dealt with in two stages, when the fireball temperature is above and when it is below the melting point of the carrier soil. In the first stage the more refractory, higher boiling, oxides of the fission product isotopes condense onto the droplets of liquid soil and diffuse into the interior of each droplet. In this form they are not biologically available for uptake by and incorporation into plants and animals, when the solidified particles reach the ground. During the second stage, as the fireball cools through their successive boiling points, the other fission product isotopes or their compounds condense on to the surfaces of the solidified carrier soil particles. Since larger particles fall earlier out of the rising fireball, those fission products of lower boiling points condense later on to smaller particles which are carried further downwind with increasing biological availability to plants and animals (p.77 shows solubility - particle size relationships for underground, tower and balloon bursts at Nevada). Those fission product isotopes which are permanent gases or have gaseous precursors tend to escape from the fireball and cloud.

C. Fractionation of fission products

The general effect of the condensation process can be seen in Fig. 6.2 p.320 based on weapon trials (p321), in which $T_\alpha(1)$ is plotted against the particle size-location parameter α which increases with downwind distance: $T_\alpha(1)$ is the ratio of the H + 1 hour radiation intensity at the 3 ft. level over (actual) fractionated to unfractionated fission products per fission per sq. foot at the location corresponding to the value of α. Fig. 6.2 shows that even at great distances from the point of burst i.e. for $\alpha > 100$, there will always be missing about 20% of the products from each fission, escaping as gases from the fireball and cloud.

In Miller's model, the melting point of the soil material is arbitrarily chosen as 1400°C. The time for the fireball to cool to the

solidification temperature, (in this case 1400°C) is critical and also increases with yield. The temperature of the second maximum for the fireball of a groundburst is given on p.141 as

$$t_2 = 0.61\ W^{0.373} \text{ secs.}$$

The fireball Temperature T°K is found to vary with time as $(t/t2)^{\frac{1}{3}}$ for values of t/t_2 from 1 to 10 and as exp $(-kt/t_2)$ at later times (p.143 presumably determined from weapon trials). This leads to equations 3.133 and 3.135 on p.143 expressing T as a function of yield and time. Putting in the value T = 1673°K for the melting point of the soil and also the arbitrarily selected times of 9 and 60 secs into these equations, Miller finds that the fireballs from 84 KT and 14 MT groundbursts would cool to 1400°C respectively in these times of 9 and 60 seconds.

Miller's arguments and calculations of fractionation factors from his mathematical model and those deduced from the radiochemical analysis of fall-out samples collected at weapon trials are not easy to follow because of the many symbols used for different ways for expressing fractionation factors e.g.

$$\gamma_\delta A(t), \quad \gamma_\delta' A(t), \quad r_{fp}(\bar{y}), \quad r_\alpha(t) \text{ and } r_x(t)$$

(all of which vary with time t) and also because it is difficult to fit into the picture the activity lost in very small world-wide fall-out particles as this is to some extent independent of fractionation of the products from single fissions and may be regarded more as fission losses.

Miller uses the basic data of Bolles, Ballou and others Table 2.3 (pp.20-22) on the abundances of fission products from the fission of U235 by thermal and by fission neutrons, from the fission of U238 by 8 Mev neutrons and from the fission of Pu239 by fission neutrons and their decay rates (Table 3.17 pp. 178 to 182 and Fig. 3.5 p.185). He calculates (Table 3.15 p.169) for each nuclide in each mass chain (excluding only those which disappear before 45.8 mins. or 0.763 hours), the fraction $r_\delta A$ which would condense up to periods of 9 secs and 60 secs corresponding to the soil solidification times for fireballs of 84 KT and 14 MT. groundbursts. These data are converted to normalised air ionisation rates in Tables 3.16 pp. 171-176 i.e. r.p.h. per disintegration per sec. per 10^4 fissions per sq. ft: both absolute values, "metered" values which would be registered by a man and radiac instrument with a shielding factor of about 0.75 are given. The ionisation rates for the fractionated individual fission products per fission per sq. ft. are then summed relative to the totals for unfractionated fission products: the values for the latter at H + 1 hour are shown in Table 3.18 p.187 for different types of fission (U235, U238 and Pu239). The calculated ratios r_{fp} for fractionated to unfractionated fission products for 84 KT and 14 MT groundbursts from U238 fission by 8 Mev neutrons at 45.8 mins. (0.763 hours) and during the subsequent decay is shown in Fig. 3.10 p.193 (in this case relative to unfractionated fission products from U235). As indicated in the text on p.190 the first depression in both curves at about H + 2 hours is due to major depletions in C_s and T_e and those at about 200 hours to depletions in I^{131} and Ba/La^{140}. Fig. 3.10 should be compared with Fig. 3.11 p.195 which shows how the ratio r_{fp} varies with time, based on measurements from low tower shots in Nevada. Fig. 3.11 also indicates, on the abscissa, the elements in high abundance at various times in the fission products. The general trend of the curve is similar to that for the 60 sec fireball cooling time to the soil melting point.

Continuing his theoretical study Miller next considers what additional fractions of the fission products would condense on to soil particles

after they had solidified: he extends the time for this second stage of condensation to 406 secs. by which time $> 6\frac{1}{2}$ minutes the cloud would become stabilised. The data for the individual isotopes of each mass chain A expressed as the fraction $r_o(A)$ in Table 6.1 (pp.307-310) are additional to those values of $r_o(A)$ in Table 3.15 (p.169) for the first condensation stage. The selected intermediate times 238 and 174 secs. represent respectively the mid-time points for the cloud expansion of 84 KT and 14 MT groundbursts. Comparison of the two indicates that the fission product condensation is complete by about 200 secs. for all except the isotopes of Se, As and the permanent gases.

The effect, of assuming that different limiting times from 9 secs. out to 406 secs. are available for the condensation of fission products from 84 KT and 14 MT groundbursts, for both U235 fission by thermal neutrons and U238 fission by 8 Mev neutrons, is shown by the absolute air ionisation rates i_o and the fractionation factors $r_\alpha(1)$* at H + 1 hours in Table 6.2 p.311 and the corresponding decay data from 0.763 hours in Table 6.3 p.313 also presented graphically in Fig. 6.1 pp. 314-315 as air ionisation rates per fission per sq. ft. Miller concludes on p.311 that in local fall-out the gross fraction of the ionisation rate at 1 hour has a maximum value of about 0.8 and that no large effect of yield is indicated. On p.253 however he points out that, at least for larger yields where the fraction of the activity on the ground but beyond the 1 rph at 1 hour contour is very small, both the overall fractionation and the fraction contributing to world wide fall-out (i.e. the lost fissions) decrease with increasing yield, because of the slower cooling of the fireball.

Miller concludes on pp. 312 and 319 that the best agreement between calculated and observed $r_\alpha(t)$ data is obtained by assuming a condensation delay time of 180 secs. in the fireball. This means that the larger particles which fall-out of the fireball much before 180 secs. and will have smaller values of α, will suffer considerable depletion in some of the fission products. Those particles which fall-out of the cloud after 180 secs. will have fractionation factors approaching the limit of 0.8 representing depletion only of permanent gas fission products.

D. Experimental Data on Fractionation

Chapter 2 reviews the available published data on the characteristics of fall-out particles, including radiochemical analysis in samples taken at various weapon trials and Miller gives, mainly in Chapter 3, such fractionation factors as can be deduced from the experimental data (e.g. from the Nevada tower shot Fig. 3.11 p.195 already mentioned in these notes). Unfortunately the American and British published radiochemical data are inadequate for the reliable calculation of the fractionation factors of individual isotopes in close-in fall-out consisting of particles larger than 50 microns diameter.

The most valuable data, are the analyses of relatively close-in fall-out particles in the large samples taken from the Japanese ship No 5 Fukuryu Maru after the 14 MT Bravo Coral shot in 1.3.54. Kimura (see p.50 and ref. 23 on p.94) reporting on this work, assumed that the fission

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* The use of $r_\alpha(1)$ in this context is rather confusing and it might have been better to use the symbol $r_{fp}(1)$: but the comment on p.312 about the applicability of the symbol $r_\alpha(t)$ to the fractionation factors is in Table 6.14 (p.318) is relevant.

products came from Pu239 and Miller recalculated the data to U238 fission by 8 Mev neutrons. He summarises, in Table 2.6 p.54, for a number of individual beta - emitting isotopes, in samples of fall-out at D + 21 and D + 25 days, the fractionation factors $r_o(A)$ relative to the unfractioned ones (e.g. Zr^{95} and Mo^{99}) after allowing for induced activity due to neutron capture by U238, estimated from Kimura's report on the ratio of Pu239 and U237 activity to that of the fission product samples, at 0.3 atoms of Pu239 and 0.15 atoms of U237 per fission. On p.56 however he queries the Japanese alpha counting technique and indicates that there is some evidence for assuming the capture by U238 of at least 0.75 neutron per fission and in his mathematical model he takes 0.8 neutrons per fission (p.231)(and 1 on p.196)

Other sets of less complete data on fractionation are reviewed in pp. 56-65 and some interesting general conclusions on fractionation are given on pp. 62 and 63 while the build-up and decay, during the first 100 seconds of the various isotopes in each of the mass chains 89, 90, 137 and 140 are illustrated in Fig. 2.12 on p.64. The differences between fractionation factors for some fission isotopes relative to Zr^{95}, in balloon and tower shots are shown in Table 2.12 p.77 with an indication of relative solubility (biological availability) and particle size in Table 2.13.

E. Induced Activity (U237, U239/Np239 and U240 and Normalised Air Ionisation Rates (or dose-rates) at 3 ft. level in r.p.h. at 1 hour per KT per sq. mile

Miller deals with the activity induced in U238 on pp. 196-200: Tables 3.21 to 3.24 show for each of these possible induced isotopes, the contribution that would be made by one induced atom per fission (C = 1) to the absolute air ionisation rate Ia (or to the rate Io measured with a portable radiac meter) at 3 ft. level over an infinite smooth plane contaminated with fission products from 10^4 fissions per sq. ft. On p.196 he calculates that 1 atom of U239/Np239 per fission would add 180 rph at 1 hr. per KT per sq. mile to the air ionisation rate. Kimura's figure of 0.15 atoms of U237 per fission would only add 1 rph but 0.15 atoms of U-240 per fission would add about 20 rph at 1 hr. per KT per sq. mile to the air ionisation rate.

Since the normalised air ionisation rate for fission of U-238 by 8 Mev neutrons is given on p.187 as 3610 rph at 1 hr. per KT per sq. mile the induced activity contribution seems to be 5% for unfractionated fission products to about 15% in early fall-out where the fractionation factor may be as low as $\frac{1}{3}$ (i.e. $\frac{2}{3}$ depletion of fission product activity).

Quoting on p.187, the value given in the "Effects of Nuclear Weapons" (1957) of 1240 rph at 1 hr. per KT per sq. mile, Miller implies that this normalised value of 1240 refers to the absolute air ionisation rates when he compares it with the calculated values in Table 3.18 of 3610 for the fission of U238 by 8 Mev neutrons. On the other hand, he implies on p.196 that the normalised value of 1240 refers to a dose-rate measured with a portable radiac meter (shielding factor 0.75) and over ground with a roughness attenuating factor of 0.75 and neglecting the relatively small proportion of induced activity this would correspond to an absolute air ionisation rate of about 2210 and a fractionation factor of 2210/3610 of about 61%. The corresponding values given by Miller's simplified mathematical model on p.296 are 2550 rph at 1 hr. per KT per sq. mile and a fractionation factor of 71%.

In a real fall-out pattern there will be other losses of fission products such as Zr^{95} and Mo^{99} which condense early in the fireball

but escape in very small particles as world-wide fall-out beyond some arbitrarily selected contour usually taken as 1 rph at 1 hr. Miller quotes on p.296 (and some data in pp. 247 and 254 are also relevant) normalised data in rph at 1 hour per KT per sq. mile calculated from various cloud models devised to reproduce actual fall-out patterns in agreement with those at weapon trials. The data on p.296 are for 10 MT, 100% fissions and they are expressed as K(1)(C1) or K(1), where C(1) is the fraction of the fissions accounted within a dose-rate contour of 1 rph at 1 hr. For a real fall-out pattern therefore Miller gives

1460 rph at 1 hr/KT/sq. mile from the ENW model
1500 " " " " " from the WSEG-RM10 model
1500 " " " " " from Anderson's model
1430 " " " " " from Miller's simplified model

The above values represent 40-43% of theoretical air ionisation rates for unfractioned fission products (3610 from U238 by 8 Mev fission or 3940 for U235 fission see p.187). On the same page (296) Miller quotes corresponding values for K(1) which would appear to take into account the fraction of world-wide fall-out containing condensed fission products that would reach the ground (but presumably still excluding those which escape as premanent gases). These values of K(1) for the different models are:-

2500 for the WSEG - RM10 model
2400 for the WSEG - NAS model
Ca 2000 for the Weather Bureau model
Ca 2550 for the Miller simplified model

F. Home Office (SAB) comparison of Miller's normalised dose-rate and those given in the 1962 edition of the Effects of Nuclear Weapons (p.492)

It is interesting for C.D. planning purposes to compare the normalised dose-rate data quoted by Miller for different cloud models and the data in Fig. 9.179 (p.492) of the latest edition of ENW. The latter shows dose-rates, at 3 ft. above an ideal plane having a contamination density of 1 gamma megacurie per sq. mile, of 6.4 rph from 0.9 Mev photons (and 5.2 for 0.7 Mev photons) corresponding to 16.6 rph per gamma curie per sq. metre.

Since we are primarily interested in U-238 fission by 8 Mev neutrons at H + 1 hours we can use the basic data of Bolles, Ballou and Glendenin, quoted by Miller on p.180, and the photon energy for U-238 in Fig. 13.8 p.189 of just over 0.9 Mev from H + 1 to H + 2 hours (dropping regularly thereafter to 0.7 Mev at H + 8 hours). Interpolation gives 1.41 photons per sec. per 10^4 fissions at H + 1 hours and since there are 1.45×10^{23} fissions/KT (Miller's p.187) we get 2.04×10^{19} photons/sec/KT at H + 1 hours. Hence a contamination density of 1 KT per sq. mile at H + 1 hours corresponds $\frac{2.04 \times 10^{19}}{3.7 \times 10^{10}}$ = 550 gamma megacuries

The ENW value of 6.4 rph per gamma megacurie/sq. mile corresponds therefore to dose-rates of 6.4 x 550 = 3520 rph at 1 hr per KT per sq. mile for U238 fission by 8 Mev neutrons and average photon energy of about 0.9 Mev. This is virtually the same as the normalised air ionisation rate of 3610 derived by Miller in Table 3.18 p.187.

It seems therefore that the data in ENW (1962) p.492 relate to absolute air ionisation rates in the sense used by Miller which thus require modification by factors for portable instrument and ground roughness shielding as well as fractionation factors of < 0.8 for fission yields under 1 MT.

For practical purposes therefore the value of 7.5 rph per (disintegration) curie per sq. metre usually accepted for CD purposes in the Scientific Adviser's Branch, Home Office, seems more than adequate for MT groundbursts on ground with a combined roughness factor and instrument factor of 0.5.

Appendix

List of some misprints and errors in "FALLOUT AND RADIOLOGICAL COUNTERMEASURES" VOLUME I

P.63 6 lines from bottom, for "gass" read "gas"

P.69 Para. 1, line 7, for raionuclides" read "radionuclides"

? P.81 Line 12, "Wind Vector = wind speed x time"

P.82 "Wind vector (miles) = wind speed x time"

P.100 3 lines from bottom, for "n", read "n_j"

P.125 Para. 2, line 14, for "n_1" read "n_2"

P.142 Line 1, for "t = 1.4t to t = 10t" read "$t_m = 1.4t_2$ to $t_2 = 10t$"

P.147 Heading, for "VOLUES" read "VALUES"

? P.148 Last para., line 3, for "Figure 3.1", read "Figure 3.3" ?

P.157 Para. 2, line 8, for "firball" read "fireball"

P.164 (3.176), for "3.8" read "3.12".

? P.165 (3.177), for "Table 2.23" read "Table 3.10" ?

P.171 After heading, add "PER 10^4 FISSION"

P.177 Line 4, for "Section 3.4" read "Section 3.2"

P.184 Line 5, for "Figures 3.3 and 3.4" read "Figures 3.5 and 3.6"

5 lines from bottom, for "Figures 3.2, 3.3 and 3.4" read "Figures 3.4, 3.5 and 3.6"

P.187 Table 3.18 heading insert "air" after "H + 1"

P.189 Graph, change first "4" to "3" and first "6" to "5"

P.196 Para. 3, line 11, for "Figure 2.9" read "Figure 3.9"

? P.197 Table. Meaning of $\left[\frac{D_a\ (1)}{(r \times 10^9)}\right]$ not understood

P.198 4 lines from bottom for "of 10 atoms" read "of 10^4 atoms"

P.216 Line 1, for "it the falling rate" read "is the falling rate"

P.223 (4.67), for "per fiss / ft" read "per fiss /ft^2"

P.232 Line 16, for "section 5.4.1" read "section 5.1"

P.234 (5.4), insert line between numerator and denominator K_a

(5.6), for "Eq.5.43" read "Eq.4.43"

P.235 (5.9), insert line between numerator and denominator $\alpha_{2,3}$

P.259 Para. 2, for "4.75" read "4.73"

P.276 Column 2, for "t_{2_8}" read "t_z"

P.294 Line 16, for "ENW[8]" read "ENW[9]" and for "Anderson [9]" read "Anderson[10]"

? P.296 Line 20, for "K(1)C(1)" read "K(1)" ?

P.302 5 lines from bottom, for "Eq. 4.74" read "Eq. 5.1"

4 lines from bottom, for "$_x(t)$ Area" read "$1_x(1)$ Area"

P.303 (6.5), for "Table 2.19" read "Table 3.11 p.156"

P.306. 10 lines from bottom, for "tubulated" read "tabulated"

8 lines from bottom, for "$r_o(A)$" read "$r_o'(A)$"

P.312 Line 5, for "Chapter 2" read "Chapter 3"

P.325 (6.15), for "$<$" read "$>$"

(6.16), insert line between numerator and denominator

(6.17) " " " " " "